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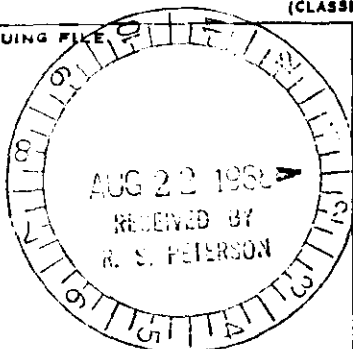
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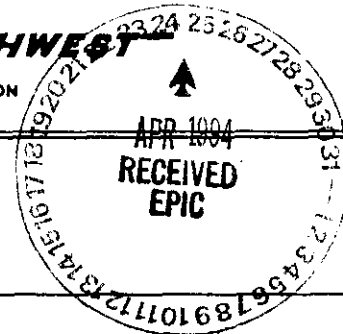
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WASTE DISPOSAL TO THE GROUND AT 100-N

AUTHOR

B. F. Hajek

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WASTE DISPOSAL TO THE GROUND AT 100-N

by

B. F. Hajek

July 31, 1968

Water and Waste Management

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WASTE DISPOSAL TO THE GROUND AT 100-N

B. F. Hajek

The objective of this document is to present information which can be used as an aid in determining the suitability of wastewater for ground disposal in the vicinity of the 100-N area. The material presented was obtained from a review of significant literature and from unpublished data from many soil-waste interaction studies conducted at Hanford.

This information is not intended for use as criteria for ground disposal but rather as a guide in decision making within the framework of criteria based on all factors which must be considered before radioactive wastewater is disposed to the soil environment.

The major sections of this document are: fundamentals, waste characteristics, and waste evaluation. The section on fundamentals discusses some theoretical concepts deemed necessary for a better understanding of radionuclide adsorption processes in soils. The section on waste characteristics is written in terms of specific radionuclides and nonradioactive competing ion effects. The waste evaluation section consists of an outline and brief explanation of how the material in this document can be used in the evaluation of suitability of wastewater for ground disposal.

FUNDAMENTALS

The factors which influence the behavior of inorganic pollutants in wastewater disposed to ground can be grouped under the general categories of hydrology and soil chemistry. The hydrologic features at 100-N have been reported in several documents (1, 2, 3, 4, 5, 6). This literature is not summarized in this report.

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Two reports on some chemical soil-radionuclide relationships in N-Area soils have been issued (7,8). Results obtained in these studies as well as many others were used to compile this document.

Many radioactive species are retained in soils by chemical reactions with constituents of soil or rock. These may be classified into two types of reactions: precipitation and ion exchange.

Precipitation

If the waste pH or the natural pH of the soil is high enough (alkaline), precipitation of hydroxides or basic salts of the transition and rare earth elements can be expected. Thus, in the neutral to slightly alkaline soil deposits found at 100-N precipitation of zirconium, niobium, lanthanum, cerium, and other elements will tend to take place. Under more alkaline conditions (pH > 8.2) some precipitation of strontium will occur. Strontium precipitation is greater if phosphates are present in the wastewater or in the soil.

Since precipitated radionuclides are retained in soils by filtration, this type of activity should remain fairly localized and generally will remain unaffected by leaching agents other than those of low pH or solutions containing complexing agents. Removal will also be largely independent of the salt content except as the ionic strength and common ions affect the solubility of precipitates. Recent studies have shown that some fine particulates (< 1 micron) can migrate through soils (7). Usually this does not present a problem if decontamination factors greater than 10^4 are not needed.

Ion Exchange

Soil exhibit ion exchange properties to varying degrees due largely to the type and amount of clay minerals present. Generally the exchange

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involves cations to a much greater extent than anions. Fortunately many long-lived radionuclides are cations and can be easily retained in soils.

In classic soil ion exchange the cation exchange capacity is a useful soil property for predictive purposes (9). However, soil exchange capacity has only limited predictive usefulness for radioactive waste disposal problems (10). The most useful soil-radionuclide inter-relationship for practical application is the equilibrium distribution coefficient (K_d) (11). This coefficient is defined by the equation,

$$K_d = \frac{\bar{C}_X}{C_X} \quad [1]$$

in which,

\bar{C}_X = activity or concentration of radionuclide X
adsorbed per gram of soil

C_X = activity or concentration of radionuclide X
per ml at equilibrium with \bar{C}_X .

Many factors effect the magnitude of the K_d . The most significant are the selectivity of soil mineral surfaces for radionuclide X, the pH of the system (some soil exchange sites are pH dependent and H^+ ions also compete), and competing cation concentrations. Usually competitive effects of other radioactive ions in the soil-waste system can be ignored in low level disposal.

The K_d for equivalent exchange can be related to the selectivity coefficient and competing cation concentration by the mass action equation,

$$K_B^X \cong K_d \frac{C_B}{C_B} \quad [2]$$

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Since \bar{C}_B is nearly constant for the soil system at 100-N ($\bar{C}_B \approx$ total cation exchange capacity), \bar{C}_B can be set equal to one and equation [2] can be written,

$$K_B^X \approx K_d C_B \quad [3]$$

The general equation for K is,

$$K_Y^X = (K_{dX.Y})^x (C_Y)^y, \quad [4]$$

where x and y are equal to the number of moles of X and Y represented in the chemical equation for exchange equilibria of X and Y,

$$x\bar{X} + yY \rightleftharpoons xX + y\bar{Y}. \quad [5]$$

Letters with the bar indicate exchanger phase (adsorbed on soil), those without bars indicate solution phase (in soil solution at equilibrium).

For disposal systems which receive pulses of waste solutions with different distribution coefficients a useful form of the K_d equation is (12),

$$K_{dx} = \frac{Q_x + \Delta X}{C_o - \Delta X \text{ (g/ml)}}, \quad [6]$$

where

Q_x = concentration of radionuclide X on the soil from previous disposal

C_o = concentration of radionuclide X in the influent solution (wastewater before disposal to ground)

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ΔX = amount of radionuclide X either adsorbed or desorbed when the soil equilibrates with the invading wastewater

ml = milliliters of solution in a volume element of soil

g = grams of soil in a volume element.

The distribution coefficient can be determined in the laboratory by batch or small column techniques or by use of equation [4] if K_B^X is known and if only one competing cation dominates. Of course the competing cation concentration also must be known. Usually the system is complicated by the presence of two or more competing ions. Mass action equations have been derived for this case (13). The general equation is

$$\sum_{i=1}^{l=n} \left[\frac{(K_{dx} \cdot 1)^x (C_I)^i}{K_I^X} \right]^{\frac{1}{i}} \approx 1. \quad [7]$$

Selectivity constants (K_I^X) for use in equation [7] are given in table 1. The constants given were obtained for solution-soil systems in which the hydrogen ion activity ranged from $10^{-7.5}$ to $10^{-6.0}$. Soil samples used include only the less than 2mm size fraction. Soils at 100-N include about 40 percent (by weight) inert gravels ($> 2mm$); consequently, all calculated K_d s should be reduced by this factor.

Equation [7] can often be simplified if it is known that only one competing ion significantly influences radionuclide retention. This is often the case for strontium-calcium and cesium-potassium systems.

It should be recognized that the constants and equation [7] cannot be used in all cases; however, this equation is often useful in the estimation of distribution coefficients when competing cation concentrations

TABLE 1
SELECTIVITY CONSTANTS FOR
TRACE Cs^+ and Sr^{++}

	<u>Na</u>	<u>Mg</u>	<u>K</u>	<u>Ca</u>
K^{Sr}	7.0	.085	6.0	0.025
K^{Cs}	160	4×10^5	2.2	8×10^5

are known (Na, K, Mg and Ca). The use of these constants and equation [7] can also indicate the need for a laboratory K_d determination when the calculated values are low and disposal suitability is doubtful; for example, a K_d less than 10 for strontium or cesium.

Radionuclide Migration

The equilibrium distribution coefficient (K_d) can be used to estimate radionuclide migration rates in soils (v_1) relative to the migration rate of the transporting solution (v). The relation of K_d to v_1 is shown in the equation,

$$v_1 = \frac{v}{1 + \frac{K_d B_d}{\theta}} \quad [8]$$

where B_d is the soil bulk density (about 1.7 for 100-N soils) and θ is the volumetric soil solution content (0.2 to 0.3). This equation describes piston flow with no dispersion and instantaneous equilibrium. A related equation,

$$CV = K_d B_d + \theta, \quad [9]$$

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in which CV is the column volumes to breakthrough, can be used if solution flow velocity is not known. A column volume is usually the volume beneath a crib to groundwater. The same assumptions apply as in equation [8]. If dispersion is symmetrical, both equations would predict the movement of the 0.5 relative concentration front. In equation [8] the average ion velocity is predicted relative to the solution flow velocity and in equation [9], the 0.5 relative concentration breakthrough is related to the volume disposed.

Many equations which relate ion migration rates, solution flow rates, and dispersion (spreading) in ion exchange columns have been derived. The predictive reliability of these equations for radionuclide movement in Hanford soil systems is not well established. The following equation is the most frequently used:

$$C/C_0 = \frac{1}{2} \left[\operatorname{erfc} \left(\frac{1 - \xi}{2\sqrt{\eta \xi}} \right) \right] \quad [10]$$

where,

C/C_0 = relative radionuclide concentration at a distance
x from the crib bottom

$$\xi = \frac{v_1 t}{x}$$

$$\eta = \frac{D}{v x}$$

t = time

D = dispersion coefficient (equal to about 0.2 cm²/hr)

$\operatorname{erfc} () = 1 - \operatorname{erf} ()$ in which erf is an error function

The dispersion coefficient (0.2 cm²/hr) was obtained from laboratory column data and has not been verified in the field for long flow paths.

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WASTE CHARACTERISTICS

The factors to be considered for ground disposal suitability will be discussed in terms of radionuclides which may be present in the waste and factors which affect adsorption. It is assumed that all radionuclides are present in trace concentrations and do not interact with other radioactive ions. That is, the presence of one radionuclide does not enhance or limit the retention of any other trace ion in the system. This is a reasonable assumption for low level wastes.

Strontium

Often strontium-90 is the radionuclide which limits crib life at Hanford. The most significant factors affecting strontium retention are pH and complementary or accompany ion concentrations (14).

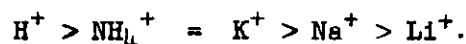
pH - The adsorption of strontium from solution by local soils is highly dependent on the reaction (pH) of the soil waste system. Essentially no adsorption occurs at pH 2; however, adsorption increases rapidly with increasing pH, especially from pH 4 to 8. Uptake reaches a maximum at about pH 10. The factors responsible for pH effects are: pH dependent charge sites, calcium and hydrogen ion competition, destruction of adsorbing surfaces, and strontium precipitation at high pH.

The pH dependent charge sites in soils are chiefly associated with edge surfaces of layer silicates, amorphous oxides, and organic matter. The cation capacity of these surfaces increases with increasing pH. Since strontium and calcium react similarly in soils, any condition which favors the dissolution of calcite (low pH) will decrease strontium adsorption. In addition, recent studies at Hanford (15) have shown that strontium is adsorbed on calcite surfaces which coat sand and silt surfaces. Above pH 8.2, calcite is essentially insoluble; however, at pH > 11 layer

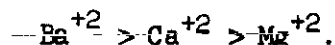
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silicate mineral solubility is increased and adsorption sites destroyed. Strontium precipitation occurs at high pH. This reaction will be discussed briefly in the next section. Consequently the most favorable pH range for strontium adsorption and retention in soils is from about 7 to 9.

Complimentary or accompanying ions - The presence of other cations in solution results in competition for adsorption sites. With the exception of hydrogen, the effectiveness of the complementary ion in reducing strontium uptake increases as the charge on the cation increases. The influence of monovalent ions on strontium adsorption is in the order,



For divalent ions the order of influence is,



Usually the competing cations encountered at Hanford are sodium, ammonium, calcium, potassium, magnesium, and of course hydrogen.

Generally, soil uptake of strontium is not seriously affected by accompanying anions. The most notable exceptions are the phosphates and oxalates. The presence of these ions usually increases strontium adsorption. Often the increase is greater than a factor of 10. It is known that in alkaline soil systems (pH > 8), a calcite-phosphate reaction occurs in which trace amounts of strontium replace a part of the calcium in the apatite mineral formed (16). A calcium oxalate precipitate also is formed in alkaline systems. Strontium could react similarly. Under certain conditions, however, oxalate and other anions may inhibit strontium uptake by soil by the formation of radiocolloids (11). Consequently it is advisable not to depend on adsorption enhancement by anions.

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Strontium adsorption on soils usually agrees with ion exchange concepts when only one competing ion is present. This is an idealized case and seldom encountered in soil-waste systems. Prediction of strontium adsorption in complex soil-waste systems by equations derived from theoretical ion exchange concepts is often difficult, especially in low ionic strength solutions such as river water. Recently statistical methods were used to develop regression equations for radionuclide adsorption on soils (17). The regression equation;

$$K_d = 20.5 - 1.7X_1 - 14.6X_2 - 3.2X_3 - 0.6X_4 + 3.1X_2X_3$$

in which,

$$X_1 = \frac{\text{ppm Mg} - 16}{10}, \quad X_2 = \frac{\text{ppm Ca} - 45}{25},$$

$$X_3 = \frac{\text{ppm K} - 13}{10}, \quad X_4 = \frac{\text{ppm Na} - 77}{75}$$

gives satisfactory estimates of the K_d for trace strontium in the presence of four competing cations. This equation is valid for all values of X between the limits of +1 and -1. The coefficients of X indicate the relative significance of these competing cations on a ppm basis. Sodium which is least significant is often the predominant competing ion. Studies at Hanford (unpublished) have shown that in river and ground water solutions sodium levels as high as 500 ppm do not seriously affect strontium adsorption at $\text{pH} > 7$.

Often concentrations of competing ions present in wastewater are not known and a reliable adsorption estimate by empirical or theoretical methods is not possible. If the waste is chemically complex it is usually better to determine actual adsorption directly in the laboratory. For rapid waste analysis the use of specific ion electrodes should be

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considered. Total cationic, sodium, calcium and total divalent electrodes are available and give acceptable concentration results in many solutions. Data obtained using cationic and anionic electrodes and conductivity data are more useful for waste behavior prediction than such waste characteristics as total dissolved solids.

In summary, the two most significant waste characteristics which affect strontium retention in soils are the pH and the competing calcium ion concentration. The most favorable pH range is from 7 to 9. The calcium ion concentration should be maintained below 40 ppm ionic calcium. Other cations such as Na, K, Mg and NH_4 usually do not limit strontium retention; however, any of these ions can limit if present in high concentrations. The question arises what is a high concentration and how can it be determined quickly? In some wastes, process data alone can provide adequate data. If more information is needed specific ion electrode and other determination methods are available. If competing ion concentration data are available, including only these ions covered in this report, the regression equation given in this section or equation [7] in the previous section can be used to estimate adsorption. For strontium the calculated K_d should be greater than 20 to allow disposal for an extended period of time with a reasonable margin of safety.

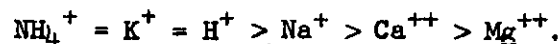
Cesium

Cesium forms relatively few insoluble precipitates and has little tendency to complex with other ions. Thus the chemistry of this radio-nuclide is comparatively simple (18). The significant characteristics to be considered for ground disposal of cesium wastes are the pH and competing cation concentration.

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pH - Cesium adsorption studies at Hanford (18) have shown that the adsorption of trace concentrations of cesium in the absence of appreciable concentrations of other cations is essentially complete at pH greater than 4. Since most wastes are neutralized because of strontium retention limitations, pH seldom limits cesium retention in soils.

Competing Ions - The ability of hydrogen and other ions to suppress cesium adsorption in equilibrium systems was found to decrease in the order,



Competing cations such as Na, Ca, and Mg can be in concentrations as high as one equivalent per liter (1 N) without seriously limiting cesium retention. Potassium and ammonium reactions in soils are similar to cesium. These ions compete for the same adsorption sites. In Hanford soil waste systems involving competition between trace cesium and potassium, cesium is slightly favored. Potassium concentrations at 100 ppm should not limit cesium adsorption in a ground disposal system which also receives strontium. The competing effects of ammonium ions can be essentially eliminated by increasing the solution pH to 10.

Generally cesium K_{ds} range from 100 to 1000 ml/g for most hanford wastes disposed to the ground.

A considerable amount of data has been published which suggests that a portion of cesium retained in soils is irreversibly sorbed.

Studies at Hanford indicate that cesium does not leach readily once it is adsorbed; however, this seems to be due more to high selectivity and not irreversibility.

Cobalt

In neutral to alkaline systems cobalt is retained in soils by filtration of the precipitated form if it is not complexed. Complexed

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anionic or nonionic cobalt is often found in wastes. These forms will not be retained.

Soil particulate retention can usually be estimated quickly in the laboratory by filtration through a millipore filter ($> 0.1 \mu$) or high speed centrifugation. Cobalt remaining in the filtrate or supernate will usually not be retained in soils.

Since precipitation is the principal retention mechanism, pH is usually the critical waste parameter. Competing cations will affect cobalt only as they affect solubility of the precipitate forms. Accompanying anions may be significant if cobalt complexes are formed.

Because of the complexity of cobalt chemistry, some laboratory analyses usually are required before cobalt soil retention predictions can be made.

Zirconium

Zirconium uptake in soils is virtually complete above pH 2. The uptake of trace zirconium-95 is not appreciably different from that in the presence of a carrier, indicating that precipitation is probably the retention mechanism (11).

Zirconium adsorption is not affected by the presence of other cations except at low pH ($\text{pH} < 2$). In these acid systems zirconium is probably ionic and consequently is more subject to competition from other cations.

Anions such as carbonates, oxalates and citrates are known to inhibit zirconium retention over a wide pH range. These and other anions, especially those of weak organic acids are believed to react with zirconium to form complex anionic species which are not sorbed. Whenever

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the presence of a complexing anion is suspected, laboratory tests, in which the wastes are filtered through small soil columns, are needed.

Iodine

Soils have essentially no iodine retention capacity. It is usually adequate to assume that this ion will migrate at the same rate as the transporting solution. This assumption allows a small margin of safety as Eliason (6) has shown that the travel time for ^{131}I is about 1.28 times less than waste water disposed to the 1301-N crib. This degree of hold-up indicates a distribution coefficient of about 0.06 ml/g.

Phosphorus

Many soils have some capacity for phosphate retention. This capacity is probably not due to ion exchange. Other mineral reactions seem to be more significant. Studies at Hanford have shown that the ^{32}P distribution coefficients for a 100 area soil is about 1.5 ml/g (19). This indicates that ^{32}P should migrate at about 1/8 the rate of a transporting soil solution which is similar to river water in ionic composition and ionic strength. Results obtained from a reactor coolant disposal test at 100-D (20), were in agreement with laboratory results.

Chromium

Little data is available on ^{51}Cr soil interrelationships. The data available show that retention of this radionuclide in soils from alkaline low ionic strength solutions is intermediate between ^{131}I and ^{32}P .

Zinc

The retention of zinc is essentially complete in alkaline systems. This radionuclide is retained as hydroxide, phosphate, carbonate or

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silicate precipitate. Little movement of retained zinc should occur unless a solution containing complexing agents like EDTA or dithizone are introduced into the system.

Chemical Wastes - Nonradioactive

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The disposal of nonradioactive wastes could reduce crib life of a facility which previously received radionuclides. Disposal of radioactive wastes to a facility which previously received only chemical wastewater is not as critical. The best approach to the evaluation of these wastes for disposal is to assume that the waste contains the radionuclides previously adsorbed and use the equations and constants given in this report. If the waste is complex (uncertain components, complexing agents, etc.) laboratory tests to evaluate leaching effects may be needed. If the waste character is such that significant radionuclides would not sorb well on soil the waste is not suitable for disposal to a site which previously received radioactive wastewater.

Disposal of radioactive waste to a chemical ground waste disposal facility could be practiced if the initial wastes did not irreversibly affect the soils. Strongly acid or basic chemical wastes will cause reactions in soils which reduce radionuclide adsorption from subsequent solutions. If both types of wastes are to be disposed to a common facility, an accurate record of waste characteristics must be maintained.

It is not possible to adequately cover all possible cases which may need to be considered; however, the following outline on an evaluation approach should be similar for most cases.

I. Analysis

1. Radionuclide concentrations
2. Major competing cations, anions, complexing agents
3. Hydrogen ion activity (pH)
4. Volume
5. Particulates
6. Disposal characteristics (batch, continuous, etc.)

II. Interpretation

1. Radionuclide concentration
 - a. Waste classification
 - b. DF required
2. Major competing cations
 - a. estimate K_d
 - b. estimate travel time
 - c. effect on previously retained radionuclides
 - d. estimate DF (groundwater or river bank)

Phase I (analysis) shows some of the waste characteristics which should be considered. A record of how the waste was generated will often supply most of the needed data. Simple conductivity and specific ion electrode measurements could supply added data. If additional laboratory tests are needed it may be best to set up soil-waste adsorption tests and determine adsorption directly.

In the interpretative phase two steps are involved. First a decision could be made on radionuclide concentrations data alone. That is, concentrations could be below limits and no DF was required or concentrations could be above limits for ground disposal. If the waste is cribbable from an activity standpoint the next step is to estimate

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adsorption (K_d) by soils. This estimate can be made from data obtained in the analysis phase (use of equations or laboratory adsorption data). The K_d is used to estimate travel time to groundwater or the river. The expected DF is obtained from the travel time and half-life.

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